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Key development issues associated with compact argon fluoride waveguide lasers have been investigated in a Phase I SBIR study. Average laser power of 4 milliwatts and pulse energy of 16 microjoules have been produced. Feasibility of a compact gas supply suitable for steady-state operation of the laser at very low gas flow rates has been demonstrated. Formation of dioxygen fluoride radicals in the laser gas mixture following excitation has been observed, and optical absorption by these species has been found to limit laser energy at high pulse repetition rates. Oxygen sources in the laser gas supply have been investigated.

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Phase I Final Technical Report

COMPACT, SELF-CONTAINED ArF LASERS

I. INTRODUCTION

As a consequence of their strong interaction with most molecular species, far uv wavelengths between 190 and 250 nm find a broad array of applications. Unfortunately, these short wavelengths are relatively difficult to generate. Potomac Photonics has developed a small KrF laser at 248 nm based on electrodeless microwave discharge excitation that alleviates some of the size and cost problems associated with conventional KrF excimer sources. In the Phase I period we have explored extension of this technology to ArF at 193 nm and achieved encouraging results.

The proposed 193 nm laser is significantly smaller than the earlier KrF devices. It also will use a highly-miniaturized gas handling system that should ultimately allow hands-off operation for periods as long as 1000 hours. In this sense the laser is self-contained. To the user, the laser will appear sealed-off.

II. PHASE I OBJECTIVES

Phase I objectives were:

1. Demonstration of at least 5 mW of average power from an ArF waveguide laser.
2. Demonstration of a feasible approach to miniaturization of a gas supply system suitable for 1000 hours of operation.

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III. PHASE I RESULTS

A brief investigation of ArF operation in a commercially available KrF waveguide laser was carried out prior to Phase I and yielded 193 nm laser pulses of approximately 30 nanosecond duration and 1 microjoule energy.

Phase I work on energy scale-up began with an effort to increase the laser excitation level and thereby improve the extraction efficiency. Fluoride glass tubing of 0.35 mm bore was procured from Galileo Electro-Optics and used to construct a laser discharge tube for use in a microwave excitation system like that used in our Model GX-1000 laser. Length of the gain region in this device was approximately 40 cm, and a cross-sectional sketch of the discharge region is shown in Fig. 1.

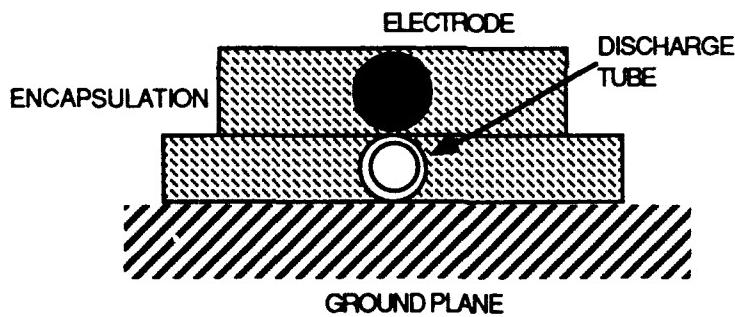


Fig. 1. Cross-section of the laser discharge structure.

Prior to the laser experiments, small-signal microwave VSWR measurements were made on a simulated discharge structure to investigate losses associated with the fluoride glass tubing. Using a slotted line set-up like that shown in Fig. 2, VSWR was measured over a frequency band between 2.5 and 3.5 GHz to determine ohmic losses associated with the discharge electrodes and dielectric losses associated with the discharge tube and potting material.

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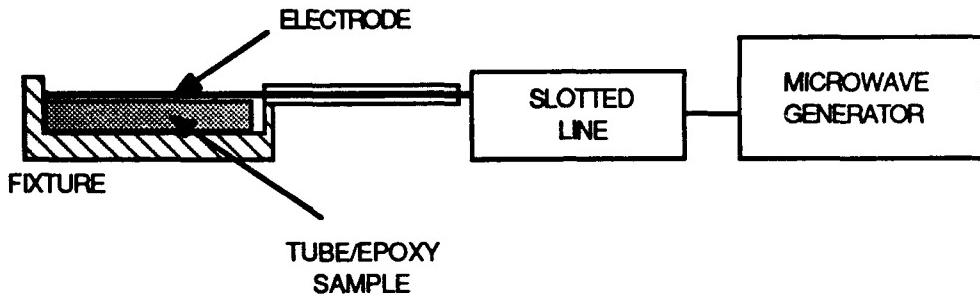


Fig. 3. Slotted line system used to measure microwave absorption in simulated discharge structures.

From the VSWR measurements attenuation coefficients were derived for microwave fields guided by the discharge structure in the absence of a discharge. These results are shown in Table I and suggest that significant losses are associated with both the discharge tube and potting compound.

<u>Configuration</u>	<u>Attenuation (%/cm)</u>
Electrode only	0.22
Electrode & tube	0.84
Electrode, tube & encapsulation	1.4

Table I. Measured attenuation coefficients for the discharge structure in the absence of the plasma.

In the Phase I laser system the discharge electrodes were approximately 15 cm long. A wave that was introduced at one end of the electrode, was reflected at the far end, and returned would experience a power loss of approximately 57% in the absence of the discharge. Microwave losses of this magnitude reduce the laser energy and efficiency. They can be reduced through use of discharge tubes with thinner walls and improved encapsulation compounds. In the Phase I study,

however, we estimate that 30 - 40% of the microwave pump energy was lost to parasitic dielectric losses in the discharge structure when the discharge is ignited.

Performance of the 0.35 mm discharge tube was initially investigated using a gas mixture of 0.2% F₂, 2.5% Ar, and balance helium (Spectra Gases, Irvington, NJ) with a 20% transmitting output coupler. ArF laser output energy was approximately 7 microjoules at repetition rates below 10 Hz decreasing to 2 microjoules at 1 kHz. At low repetition rates energy was a strong function of gas flow rate. It was found that laser energy decreased sharply when the residence time for gas in the discharge region exceeded the pulse repetition period. This behavior suggested the presence of a transient species produced by the discharge that absorbed strongly at 193 nm or interfered with the laser kinetics.

With an arrangement like that shown in Fig. 3 we used the preionization discharge that operates in a simmer mode in the center of the discharge tube as a light source to investigate transient optical absorption following the microwave excitation pulse.

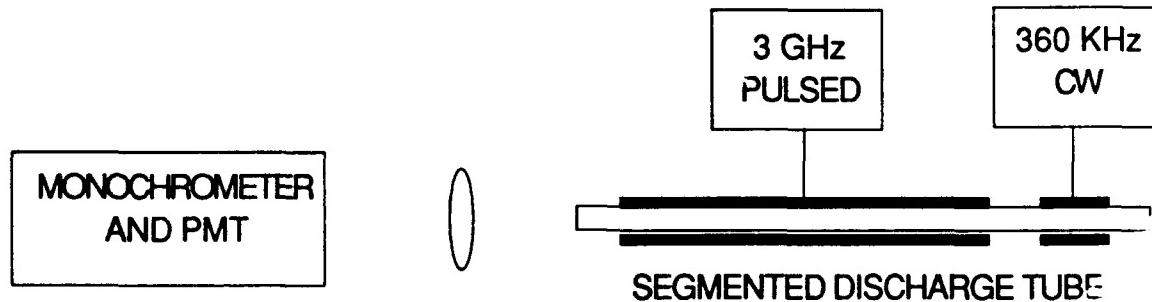


Fig. 3. Optical set-up used to measure transient absorption at 193 nm.

Figure 4 shows a typical oscilloscope trace of the 193 nm radiation transmitted through the plasma region after the

microwave pulse. Discharge-induced absorption in the 15 cm active region as high as 50 - 75% was found to exist for as much as 1 second after excitation. The magnitude of the absorption was found to decrease when gas was continuously flowed through the laser for 24 - 48 hours. Use of a liquid nitrogen cold-trap in the gas delivery line also slightly reduced the absorption. With all of these procedures, however, the peak absorption still remained near the 40% level.

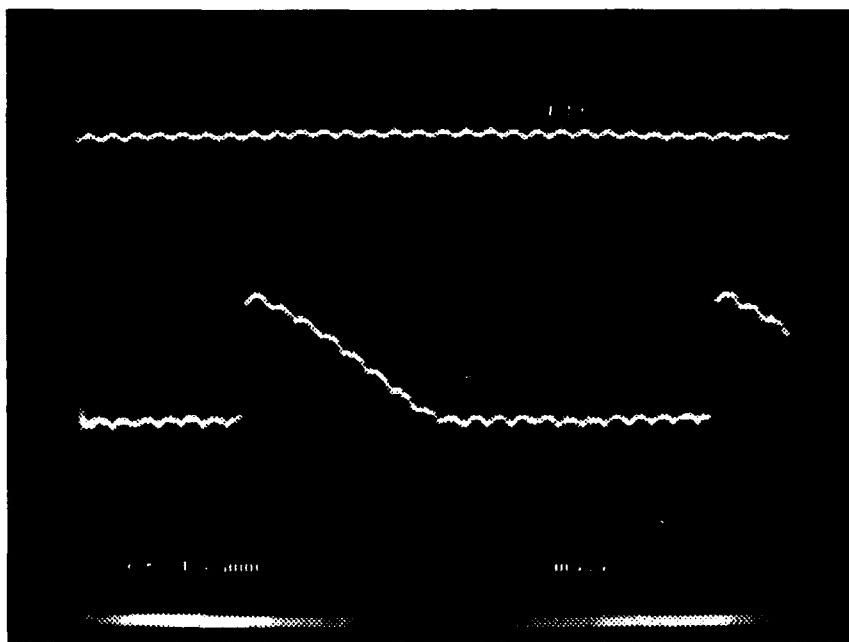


Fig. 4. Oscilloscope trace of 193 nm emission from the preionizer that is transmitted through one half of the discharge tube. The gas exchange time is approximately 160 ms. Upper trace corresponds to zero transmission. Time scale is 50 ms/div.

Efforts to identify trace contaminants in the gas that could produce the transient absorption began with uv and visible spectroscopy of the discharge plasma. Atomic and molecular spectra of low-level species in microwave discharge helium plasmas are fairly well documented[1][2]. The available data base allows fairly simple identification of

many atoms and simple molecules, although relative sensitivity varies widely. Observation of the laser

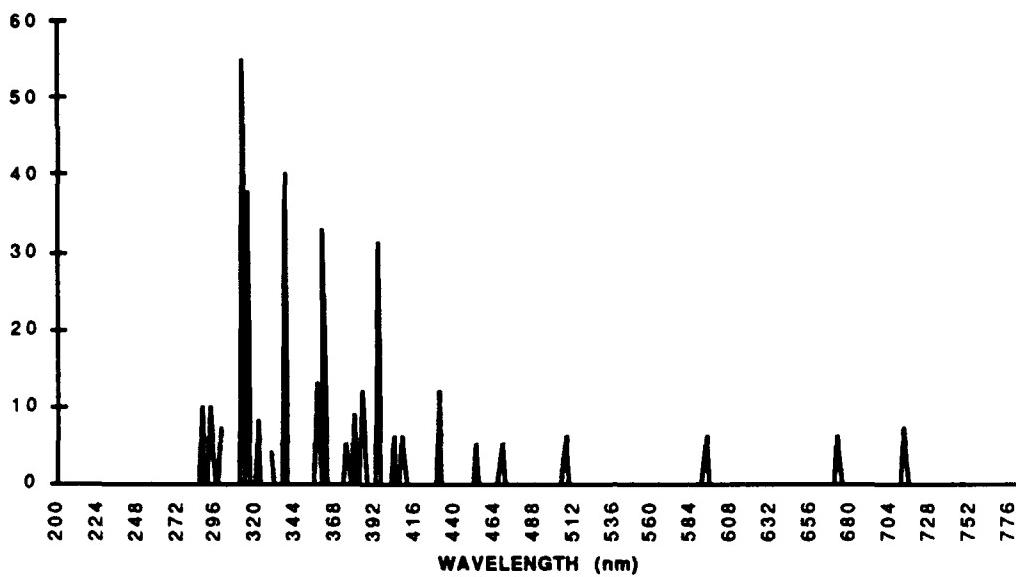


Fig. 4. Emission spectra from helium in the Phase II laser. Strong N₂ and OH emission is evident.

discharge with the fluorine and argon components removed showed strong emission bands attributable to N₂ and OH. Neither of these species absorbs at 193 nm, but several nitrogen and oxygen fluorides absorb in the deep uv. Fig. 5 shows a typical emission spectrum from a helium discharge in the laser.

Further gas analysis was carried out at Air Products Corporation at the invitation of Dr. Gene Karwacki. In this case, a small mass spectrometer was used to analyze the gas at the regulator output on the premix bottle and the gas exhausted from the laser with and without the discharge. Fig. 6 shows a mass spectrum of the gas available at the

regulator. Oxygen is seen to be one of the most prominent impurities and is found in concentrations estimated to be

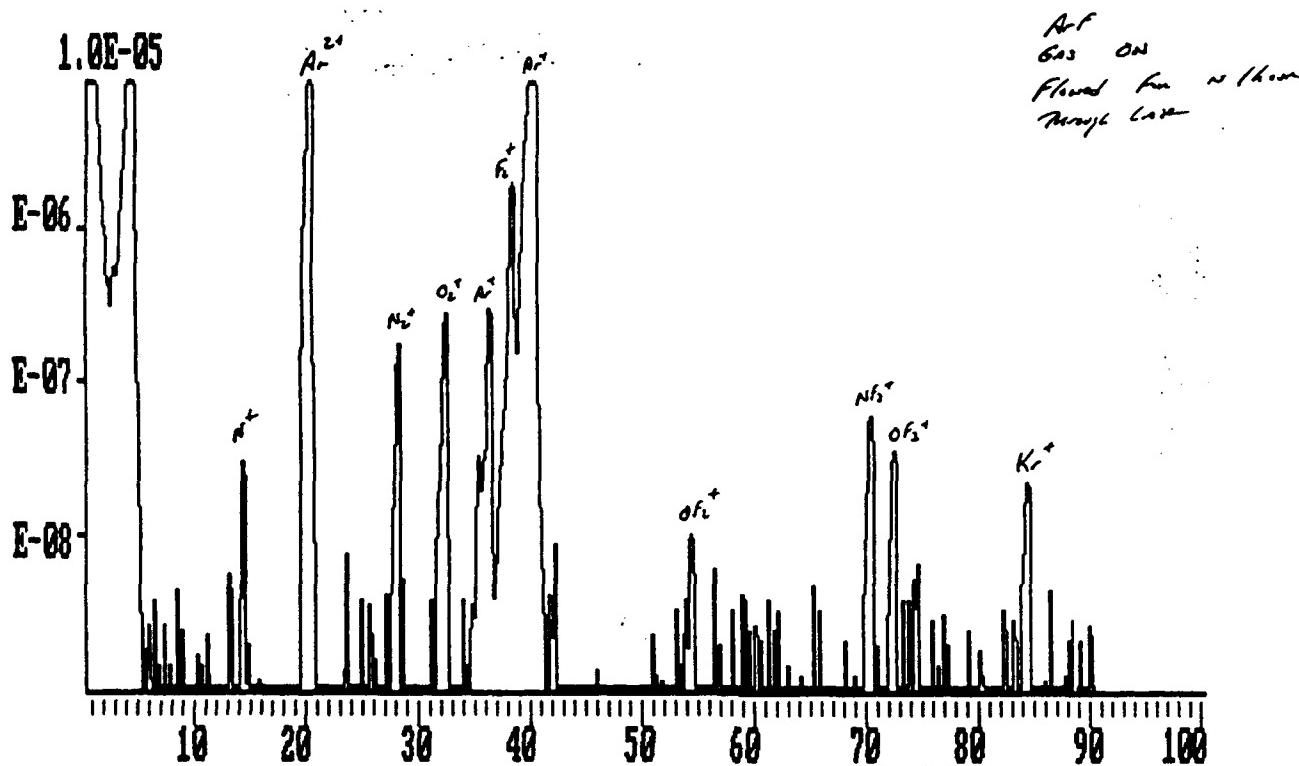


Fig. 6. Mass spectrum of ArF laser mix. An oxygen concentration of about 100 ppm is estimated by comparing the argon and oxygen peaks.

approximately 100 ppm. Spectra of nitrogen and oxygen fluorides are also evident at lower levels. Gas exhausted from the laser was found to exhibit a mass spectrum almost identical to that at the gas inlet.

With increasing evidence of the presence of nitrogen and oxygen in the laser gas, a literature search of absorption by nitrogen and oxygen fluorides was carried out with particular emphasis on transient species. It was found that O_2F and O_2F_2 are known to exhibit large absorption cross-sections at 193 nm[3]. In helium/fluorine/oxygen mixtures at atmospheric pressure, these species can exhibit a lifetime of several hundred milliseconds. They have been observed in the afterglow of a microwave plasma and as a byproduct of fluorine photolysis at 248 nm.

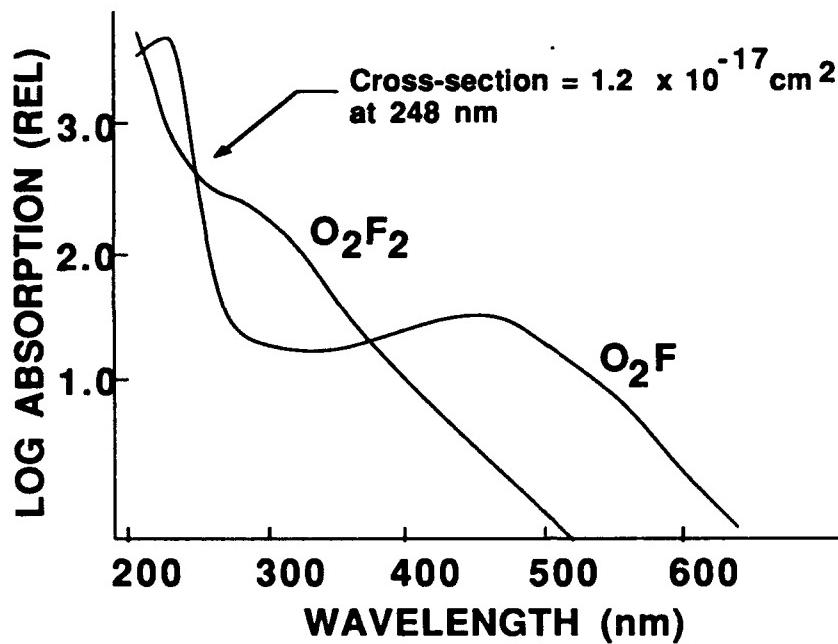
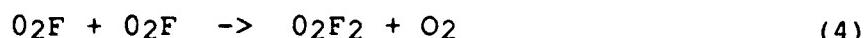
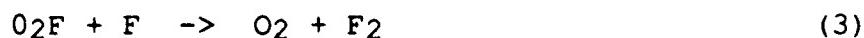


Fig. 7. Absorption spectra of O_2Fx from reference [3].

Kinetics of formation and destruction have recently been studied in detail[4]. Primary formation processes are:



Other reactions that may play a role are:





The rate constants for reactions (1) and (2) are much larger than than of reaction (7) [4]. When oxygen concentrations in a typical excimer discharge exceed only tens of parts per million, processes (1) and (2) dominate reaction (7) as a pathway for fluorine recombination. Under these conditions, nearly all available oxygen would be converted to transient O_2F and O_2F_2 species with large absorption cross-sections. Using cross-sections from reference [3] we calculate that the measured 100 ppm oxygen concentration in the laser gas would give rise to a transient absorption of more than 6%/cm or about 60% in the 15 cm discharge region.

In view of these estimates, it seems very likely that the reduced energy of the laser at high repetition rate is probably due to oxygen contamination of the gas supply. To our knowledge, this is the first time that transient absorption that is apparently due to oxygen fluoride radicals has been observed in an ArF laser. This is probably because conventional ArF lasers usually exchange the gas in the discharge region between pulses.

Oxygen contamination can result from outgassing and permeation processes in the gas supply system or be due to contamination of the laser premix. Outgassing from the surfaces of supply lines and gas permeation of elastomer seals has been studied in detail, and typical rates of impurity generation are available from the literature. Water vapor is typically the predominant impurity produced by outgassing with lower concentrations of atmospheric gases and volatile hydrocarbons. Permeation of elastomer seals usually introduces atmospheric gases into the laser mixture. We have made quantitative estimates of impurity levels that should be expected from the various sources. For outgassing

and permeation, the impurity level is a function of the gas flow rate, the surface area of the delivery system, the materials of construction, and the thickness of seals. Fig. 8 shows estimated impurity concentrations as a function of gas flow rate for various materials of construction in a gas system comparable to that used for the Phase I laser. In this case most of the surface area lies in the gas regulator and the mirror seal areas. The mirror seals themselves are Teflon encapsulated Viton. Almost any impurity present in the laser gas at concentrations of several hundred ppm will reduce laser output through interference with laser kinetics. Impurities that photodissociate to form deposits on laser mirrors can cause long-term problems even at concentrations in the few ppm range.

Back diffusion of atmospheric gases through leaks is another source of impurity introduction. Small leaks can be difficult to detect, but can be an important source of contamination. During the transient absorption measurements, we observed a substantial increase in absorption when one of the mirror seals was not properly seated and produced a small leak to ambient atmosphere.

Another potential source of contamination is impurities in the laser premix gases. All of the rare gases can be obtained with very high purity. Fluorine, however, is available with purity of only 98 - 99%. This is a potential problem in ArF systems. In Phase I, laser fluorine concentrations were typically several tenths of a percent, so that impurity levels of 50 - 150 ppm are probably introduced through the fluorine component of the gas. Comparison with Fig. 8 suggests that in a leak-free vacuum system constructed from materials with low outgassing rates the fluorine source is likely to be the dominant source of impurities.

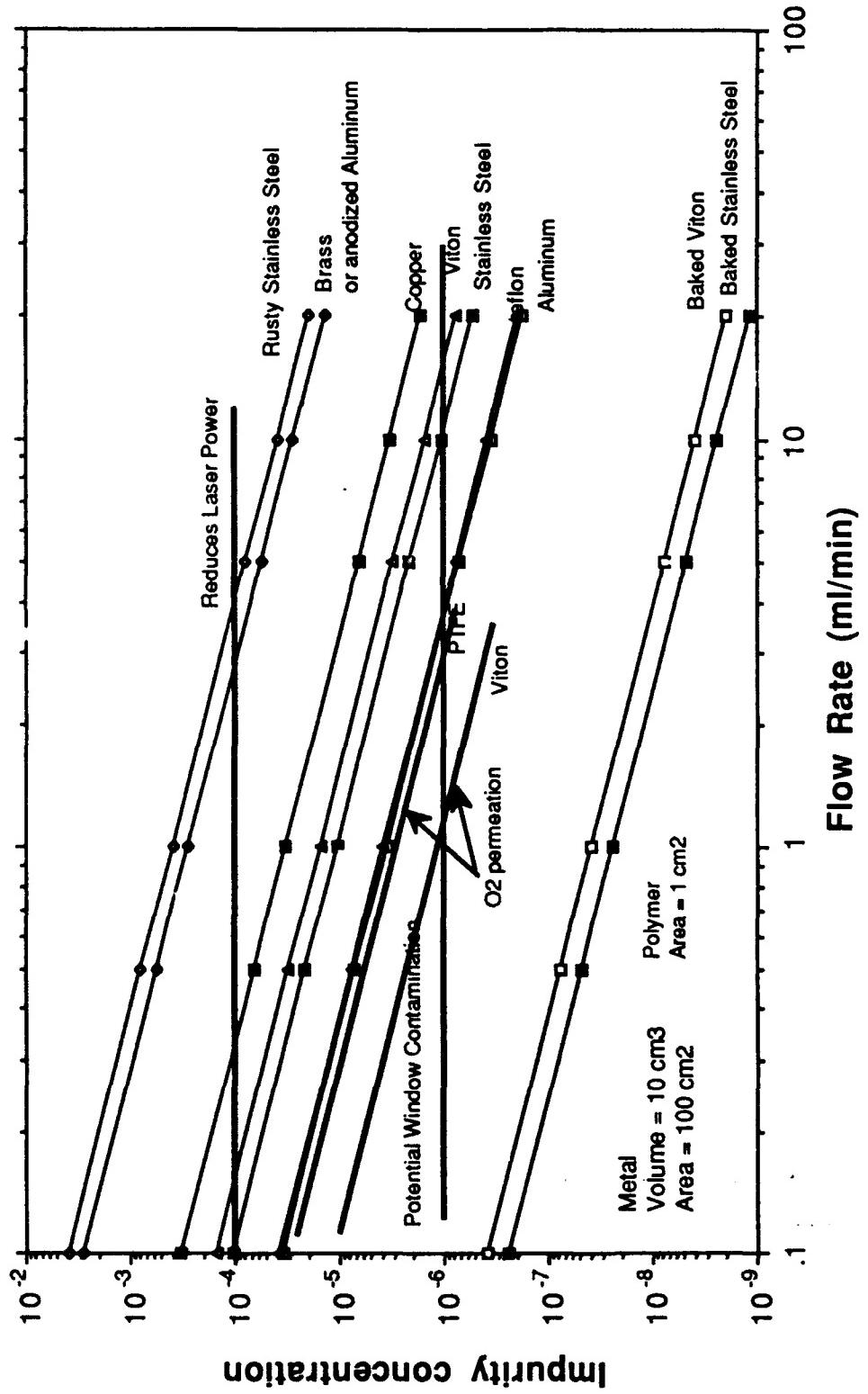


Fig. 8. Estimated impurity concentrations in the ArF laser gas due to outgassing for various materials of construction. Also shown are impurity levels for permeation of seals made of Viton and Teflon. It is assumed that all of the materials have been in a high vacuum environment for a period of 10 hours prior to use. Note that materials that tend to form thick oxides and polymers tend to outgas extensively. Impurity levels that can produce quenching of the laser species or cause photodeposition on the mirrors are indicated.

Air Products is one of the few fluorine manufacturers in the U.S. Dr. Karwacki at Air Products describes the composition of fluorine impurities as predominantly O₂, N₂, and CF₄. These compounds are difficult to separate from fluorine by conventional distillation processes, but can be removed by specially designed stills and selective absorption processes. Air Products has recently set up a still to produce high purity fluorine, but this has only recently come on line. Discussions with Spectra Gases reveal that they use fluorine from Air Products in their laser premixes and are currently working on a proprietary process to purify fluorine to the 99.99% level. They have developed highly sensitive GC/MS techniques for measurement of low-level impurities in excimer gas mixture. They confirm that oxygen concentrations at the 100 ppm level are likely to be present in their laser premixes, but expect higher purity mixtures to be available by mid-1992. During the course of the Phase I experiments, however, there appeared to be little recourse to use of relatively impure fluorine sources.

Keeping in mind the limitations imposed by available gas sources, we carried out a program for development of a high-purity delivery system and reduction of impurity sources in the laser head. A 316L stainless steel lecture bottle was fitted with a stainless steel diaphragm valve and a low-internal-volume electropolished stainless steel regulator. This combination was used as the storage and pressure regulation components of the gas delivery system. VCR fittings were used throughout. High purity nickel tubing of 1/16" OD with a smooth inner bore was procured for use as delivery line. The tubing was cleaned with acetone and alcohol rinses, dried at 150°C with helium flow and passivated with a flow of 1% F₂ in helium. A small oven was constructed and the entire storage tank, valve, and regulator assembly was baked at 100°C for 48 hours with constant flow of helium and fluorine/helium mixtures.

In the laser head, the primary sources of contamination are the mirror o-ring seals and the epoxy seal at the discharge tube. During Phase I we investigated two alternatives to the mirror o-ring seals that are much less susceptible to outgassing and permeation. These flexible metallic seal configurations use a Belleville disc spring or a metal c-ring to allow the mirror to be tilted for alignment without need for an elastomer seal. An indium layer is used at the sealing surfaces to assure a tight seal. The Belleville spring approach is desirable since it allows a wider range of adjustment with less mechanical stress on the mirror surface. We also have developed a hard sealing technique in which the gas seal at the end of the discharge tube is initially made with epoxy, but is later covered with a layer of fluoride glass. During fabrication, a ring of the glass is bonded to the metal fixture surrounding the discharge tube, and the discharge tube is fixtured in place using epoxy. A heated tool is then used to melt the upper surface of the fluoride glass ring, causing it to flow and seal to the discharge tube.

Optimization and final testing of the Phase I laser was carried out using a tube fabricated from 0.35 mm bore fluoride glass tubing with metal c-ring seals at the mirrors. The assembled tube was heated to 80°C while purging with helium and passivating with 1%F₂/He mixture. Using an in-house gas mixing station that was carefully cleaned and passivated, gases provided by Spectra Gases, and the clean gas supply system described above we evaluated laser performance as a function of gas composition and transmission of output coupling mirrors. Highest output power was obtained with an output coupling of 40% and a 0.6%F₂, 4% Ar, 95.4% helium gas mixture. Figure 9 shows the laser pulse energy as a function of pulse repetition rate.

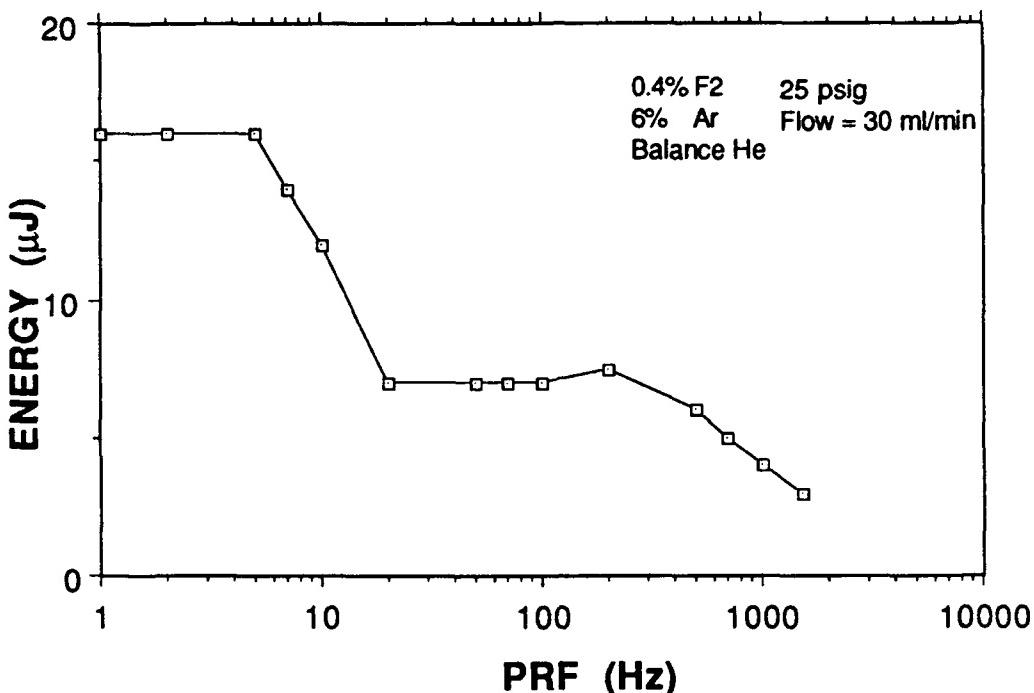


Fig. 9. ArF laser energy as a function of pulse repetition rate under optimized conditions.

The second goal of the phase I effort was demonstration of a feasible approach to a small gas supply capable of operating the laser for 1000 hours. At very low flow rates, laser output can drop significantly if impurity concentrations rise in the active region or if the halogen concentration decreases. This can occur as a result of , outgassing, permeation, or leakage that produces high impurity concentrations at low flow (see Fig. 8), chemical reactions in the gas handling system, or reactions of the discharge plasma with the discharge tube. With the advent of fluoride glass discharge tubes, the tube wall is nearly inert to the halogen plasma. Most problems at low flow are then a result of reactive materials or improper preparation of the gas system.

To demonstrate feasibility of a miniature gas supply, we needed to show that the laser could be operated in steady state with at low flow rates with a major portion of the

output energy that is available from the same system at high flow rates. In an ArF system with transient absorption due to impurities in the premix gases, large decreases in laser energy will be observed at low flow rates regardless of the quality of the gas delivery system. Consequently, the slow flow experiments were conducted with the laser operating on the KrF transition. This is a reasonable substitution, since most of the chemical processes of interest are the same for both KrF and ArF.

To investigate the effects of gas flow we set up the laser for KrF operation and inserted a diversion valve in the laser exhaust stream that allowed rapid switching from flow rates of 20 ml/min to 1 - 2 ml/min. By monitoring the changes in laser energy in response to a sudden change in gas flow rate some insight can be gained into the interaction of the gas with various parts of the delivery system. For example, fluorine reactions or outgassing from the small volume near the laser mirrors will produce an almost immediate reduction in power when flow is suddenly decreased. If the processes are occurring high-pressure side of the regulator, there will be a delay and the power will decrease with a long time constant.

With a standard stainless steel regulator on a conventional commercial gas premix tank, we found that a sudden reduction in gas flow to 1 ml/min resulted in a long slow decrease of laser energy by as much as 75%. However, with the clean 0.5 l stainless steel tank/regulator system described above we found that reduction in flow from 20 ml/min to 1.2 ml/min produced a 15% reduction in energy with no further decrease over long periods (see Fig. 10). This suggests that some improvement is needed in construction or preparation of the regulator or control valves, but the laser is in steady state operation for more than 6 hours at nearly full power with gas consumption corresponding to 1 lecture bottle per 700 hours.

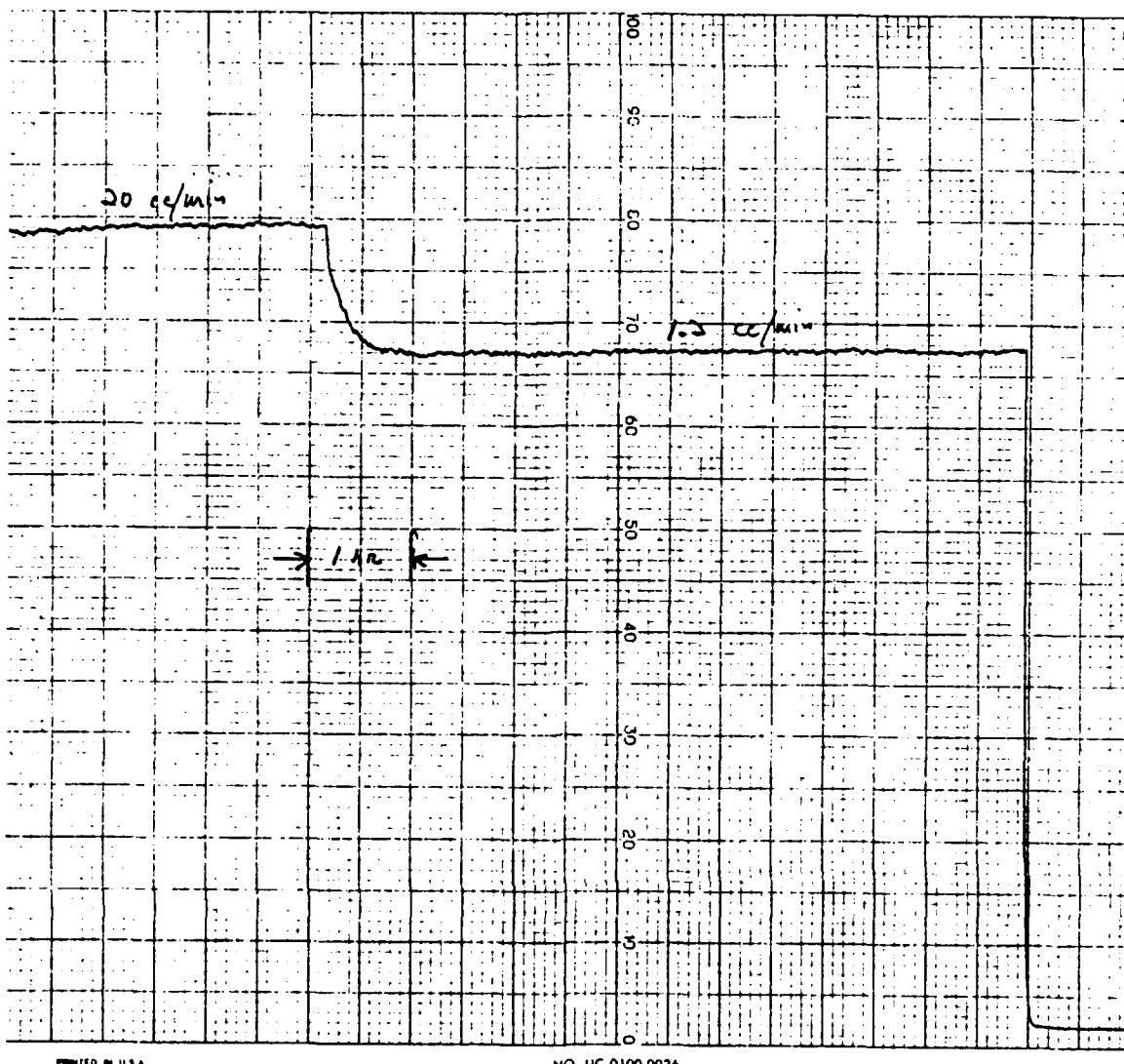


Fig. 10. Chart recording of KrF laser output power during switching from flow rate of 20 ml/min to 1.2 ml/min. The laser is in steady state operation at low flow for more than 6 hours. Laser pulse repetition rate was 500 Hz with pulse energy of 32 μ J at 20 ml/min. Chart speed is 2 div/hr.

These results clearly demonstrate feasibility of constructing the desired miniature gas supply with 1000 hour capacity. Long term storage of excimer laser premixes in steel cylinders is routinely carried out by laser gas suppliers, so that shelf life is unlikely be a problem with proper cylinder preparation. Suitable materials and effective

construction techniques for the regulator/delivery sections are demonstrated by the above experiments. Remaining is miniaturization of the regulator and valve components, which is a reasonably straightforward engineering problem.

Summary of Phase I Results.

The two Phase I objectives have been, for the most part, achieved:

- (1) An ArF waveguide has been constructed that produces energies as high as 16 μ J and more than 4 mW of average power.
- (2) A small gas supply has been constructed that will allow operation of the laser for long periods at flow rates in the 1 ml/min (1 lecture bottle/1000 hr) range.

C. Collaboration with MIT Lincoln Laboratory

Collaboration with the Submicrometer Technology Group at MIT Lincoln Laboratory was proposed as part of the Phase I work. The Lincoln Laboratory group offered use of gas and surface analysis equipment for the project, but it proved to be more expedient to use private service laboratories for the analytical work that was needed. However, the Phase I laser is being provided on loan to Submicrometer Technology Group, now headed by Dr. Mordy Rothschild, for evaluation. Trial experiments are planned involving use of the laser in interferometry or materials studies.

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